CLAIMS

We claim:

1. A method for analyzing multivariate spectral data, comprising the steps of:

a) creating a calibration model for a calibration set of multivariate spectral data **A** by:

i) obtaining a set of reference component values **C** representative of at least one of the spectrally active components in the calibration set of multivariate spectral data **A**,

ii) estimating pure-component spectra $\hat{\mathbf{K}}$ for the at least one of the spectrally active components according to $\hat{\mathbf{K}} = (\mathbf{C}^T\mathbf{C})^{-1}\mathbf{C}^T\mathbf{A} = \mathbf{C}^+\mathbf{A}$,

iii) obtaining spectral residuals $\mathbf{E_A}$ according to $\mathbf{E}_\mathbf{A} = \mathbf{A} - \mathbf{C}\hat{\mathbf{K}}$, and

iv) augmenting the estimated pure-component spectra $\hat{\mathbf{K}}$ with at least one vector of the spectral residuals $\mathbf{E_A}$ to obtain augmented pure-component spectra $\hat{\tilde{\mathbf{K}}}$; and

b) predicting a set of component values $\hat{\tilde{\mathbf{C}}}$ for a prediction set of multivariate spectral data $\mathbf{A_P}$ by:

i) further augmenting the augmented pure-component spectra $\hat{\tilde{\mathbf{K}}}$ with at least one vector representing a spectral shape that is representative of at least one additional source of spectral variation in the prediction set, and

ii) predicting the set of component values $\hat{\tilde{\mathbf{C}}}$ using the further augmented pure-component spectra $\hat{\tilde{\mathbf{K}}}$ according to $\hat{\tilde{\mathbf{C}}} = \mathbf{A}_\mathbf{P}\hat{\tilde{\mathbf{K}}}^T(\hat{\tilde{\mathbf{K}}}\hat{\tilde{\mathbf{K}}}^T)^{-1} = \mathbf{A}_\mathbf{P}(\hat{\tilde{\mathbf{K}}}^T)^+$.

2. The method of Claim 1, wherein augmenting step a)iv) is repeated until all sources of spectral variation are accounted for in the calibration set.

3. The method of Claim 1, wherein an F-test is used to select the number of vectors used to augment the estimated pure-component spectra $\hat{\mathbf{K}}$.

4. The method of Claim 1, further comprising the step of augmenting the estimated pure-component spectra $\hat{\mathbf{K}}$ with at least one vector representing at least one spectral shape representative of at least one additional sources of spectral variation in the calibration set prior to step a)iii).

5. The method of Claim 1, further comprising the step of augmenting $\hat{\mathbf{K}}$ to account for baseline variations in the calibration set prior to step a)iii).

6. The method of Claim 1, further comprising the step of augmenting $\hat{\tilde{\mathbf{K}}}$ to account for baseline variations in the calibration set prior to step b).

7. The method of Claim 1, wherein the reference component values $\mathbf{C}$ comprise concentrations of the spectrally active components.

8. The method of Claim 1, wherein the spectrally active components are selected from the group consisting of chemical specie, chemical interactions among species, physical variations, temperature variations, humidity variations, spectrometer drift, changes in spectrometers, and insertion effects.

9. The method of Claim 1, wherein the spectral residuals $\mathbf{E_A}$ are obtained from a full CLS model or a cross-validated CLS model.

10. The method of Claim 1, wherein the spectral residuals $\mathbf{E_A}$ are decomposed according to $\mathbf{E_A} = \mathbf{TP} + \mathbf{E}$, where $\mathbf{T}$ is a set of n x r scores and $\mathbf{P}$ is a set of r x p loading vectors obtained from factor analysis of the spectral residuals $\mathbf{E_A}$, and $\mathbf{E}$ is a set of n x p random errors and spectral variations not useful for prediction.

11. The method of Claim 10, wherein the $\mathbf{P}$ loading vectors comprise the at least one vector of the spectral residuals $\mathbf{E_A}$ used to augment the estimated pure-component spectra $\hat{\mathbf{K}}$ in step a)iv).

12. The method of Claim 10, wherein the factor analysis method comprises principal components analysis.

13. A method for analyzing multivariate spectral data, comprising the steps of:

a) creating a calibration model for a calibration set of multivariate spectral data **A** by:

i) obtaining a set of reference component values **C** representative of at least one of the spectrally active components in the calibration set of multivariate spectral data **A**,

ii) estimating pure-component spectra $\hat{\mathbf{K}}$ for the at least one of the spectrally active components according to $\hat{\mathbf{K}} = (\mathbf{C}^T\mathbf{C})^{-1}\mathbf{C}^T\mathbf{A} = \mathbf{C}^{+}\mathbf{A}$,

iii) obtaining spectral residuals $\mathbf{E_A}$ according to $\mathbf{E_A} = \mathbf{A} - \mathbf{C}\hat{\mathbf{K}}$,

iv) decomposing the spectral residuals $\mathbf{E_A}$ according to $\mathbf{E_A} = \mathbf{TP} + \mathbf{E}$ where **T** is a set of *n* x *r* scores and **P** is a set of *r* x *p* loading vectors obtained from factor analysis of the spectral residuals $\mathbf{E_A}$, and **E** is a set of *n* x *p* random errors and spectral variations not useful for prediction,

v) augmenting the set of reference component values **C** with at least one vector of the **T** scores to obtain a set of augmented component values $\tilde{\mathbf{C}}$, and

vi) estimating augmented pure-component spectra $\hat{\hat{\mathbf{K}}}$ according to $\hat{\hat{\mathbf{K}}} = (\tilde{\mathbf{C}}^T\tilde{\mathbf{C}})^{-1}\tilde{\mathbf{C}}^T\mathbf{A} \approx \tilde{\mathbf{C}}^{+}\mathbf{A}$; and

b) predicting a set of component values $\hat{\hat{\mathbf{C}}}$ for a prediction set of multivariate spectral data $\mathbf{A_P}$ according to $\hat{\hat{\mathbf{C}}} = \mathbf{A_P}\hat{\hat{\mathbf{K}}}^T(\hat{\hat{\mathbf{K}}}\hat{\hat{\mathbf{K}}}^T)^{-1} = \mathbf{A_P}(\hat{\hat{\mathbf{K}}}^T)^{+}$.

14. The method of Claim 13, wherein the augmenting step a)iv) is repeated until all sources of spectral variation are accounted for in the calibration set.

15. The method of Claim 13, wherein an F-test is used to select the number of vectors used to augment the estimated pure-component spectra $\hat{\mathbf{K}}$.

16. The method of Claim 13, further comprising the step of augmenting the estimated pure-component spectra $\hat{\mathbf{K}}$ with at least one vector representing a spectral shape representative of at least one additional source of spectral variation in the calibration set prior to step a)iii).

17. The method of Claim 13, further comprising the step of augmenting $\hat{\mathbf{K}}$ to account for baseline variations in the calibration set prior to step a)iii).

18. The method of Claim 13, further comprising the step of augmenting the augmented pure-component spectra $\hat{\tilde{\mathbf{K}}}$ with at least one vector representing a spectral shape representative of at least one additional source of spectral variation in the prediction set prior to step b).

19. The method of Claim 13, further comprising the step of augmenting $\hat{\tilde{\mathbf{K}}}$ to account for baseline variations in the calibration set prior to step b).

20. The method of Claim 13, wherein the reference component values **C** comprise concentrations of the spectrally active components.

21. The method of Claim 13, wherein the spectrally active components are selected from the group consisting of chemical specie, chemical interactions among species, physical variations, temperature variations, humidity variations, spectrometer drift, changes in spectrometers, and insertion effects.

22. The method of Claim 13, wherein the step a)iv) comprises augmenting the set of reference component values **C** with a set of random numbers.

23. The method of Claim 13, wherein the spectral residuals $\mathbf{E}_\mathbf{A}$ are obtained from a full CLS model or a cross-validated CLS model.

24. The method of Claim 13, wherein the factor analysis method comprises principal components analysis.

25. A method for analyzing multivariate spectral data, comprising the steps of:

a) creating a calibration model for a calibration set of multivariate spectral data $\mathbf{A}$ by:

i) obtaining a set of reference component values $\mathbf{C}$ representative of at least one of the spectrally active components in the calibration set of multivariate spectral data $\mathbf{A}$,

ii) estimating pure-component spectra $\hat{\mathbf{K}}$ for the at least one of the spectrally active components according to $\hat{\mathbf{K}} = (\mathbf{C}^T\mathbf{C})^{-1}\mathbf{C}^T\mathbf{A} = \mathbf{C}^+\mathbf{A}$,

iii) estimating a set of component values $\hat{\mathbf{C}}$ using the estimated pure-component spectra $\hat{\mathbf{K}}$ according to $\hat{\mathbf{C}} = \mathbf{A}\hat{\mathbf{K}}^T(\hat{\mathbf{K}}\hat{\mathbf{K}}^T)^{-1} = \mathbf{A}(\hat{\mathbf{K}}^T)^+$,

iv) obtaining component residuals $\mathbf{E_c}$ according to $\mathbf{E_c} = \hat{\mathbf{C}} - \mathbf{C}$;

v) augmenting the set of reference component values $\mathbf{C}$ with a vector of the component residuals $\mathbf{E_c}$ to obtain a set of augmented component values $\tilde{\mathbf{C}}$, and

vi) obtaining augmented pure-component spectra $\hat{\tilde{\mathbf{K}}}$ from the set of augmented component values $\tilde{\mathbf{C}}$ according to $\hat{\tilde{\mathbf{K}}} = (\tilde{\mathbf{C}}^T\tilde{\mathbf{C}})^{-1}\tilde{\mathbf{C}}^T\mathbf{A} = \tilde{\mathbf{C}}^+\mathbf{A}$; and

b) predicting a set of component values $\hat{\tilde{\mathbf{C}}}$ for a prediction set of multivariate spectral data $\mathbf{A_P}$ according to $\hat{\tilde{\mathbf{C}}} = \mathbf{A_P}\hat{\tilde{\mathbf{K}}}^T(\hat{\tilde{\mathbf{K}}}\hat{\tilde{\mathbf{K}}}^T)^{-1} = \mathbf{A_P}(\hat{\tilde{\mathbf{K}}}^T)^+$.

26. The method of Claim 25, further comprising repeating the following steps at least once prior to step b):

a) estimating augmented component values $\hat{\tilde{\mathbf{C}}}$ according to $\hat{\tilde{\mathbf{C}}} = \mathbf{A}\hat{\tilde{\mathbf{K}}}^{\mathrm{T}}(\hat{\tilde{\mathbf{K}}}\hat{\tilde{\mathbf{K}}}^{\mathrm{T}})^{-1} = \mathbf{A}(\hat{\tilde{\mathbf{K}}}^{\mathrm{T}})^{+}$ using the augmented pure-component spectra $\hat{\tilde{\mathbf{K}}}$ from step a)vi),

b) calculating updated estimated component values $\hat{\mathbf{C}}'$ consisting of the estimated component values in $\hat{\tilde{\mathbf{C}}}$ corresponding to the known reference component values in $\mathbf{C}$,

c) obtaining new component residuals $\mathbf{E_c}'$ according to $\mathbf{E_c}' = \hat{\mathbf{C}} - \mathbf{C}$, and

d) further augmenting the set of augmented component values $\tilde{\mathbf{C}}$ with a vector of the component residuals $\mathbf{E_c}'$ to obtain a new set of augmented component values $\tilde{\mathbf{C}}$ to be used to obtain new augmented pure-component spectra $\hat{\tilde{\mathbf{K}}}$ in step a)vi).

27. The method of Claim 26, wherein an F-test is used to select the number of times that steps a) through d) are repeated.

28. The method of Claim 25, further comprising the step of augmenting $\hat{\mathbf{K}}$ with at least one vector representing a spectral shape that is representative of at least one additional source of spectral variation in the calibration set prior to step a)iii).

29. The method of Claim 25, further comprising the step of augmenting $\hat{\mathbf{K}}$ to account for baseline variations in the calibration set prior to step a)iii).

30. The method of Claim 25, further comprising the step of augmenting $\hat{\tilde{\mathbf{K}}}$ with at least one vector representing a spectral shape that is representative of at least one additional source of spectral variation in the prediction set prior to step b).

31. The method of Claim 25, further comprising the step of augmenting $\hat{\tilde{\mathbf{K}}}$ account for baseline variations in the calibration set prior to step b).

32. The method of Claim 25, wherein the reference component values $\mathbf{C}$ comprise concentrations of the spectrally active components.

33. The method of Claim 25, wherein the spectrally active components are selected from the group consisting of chemical specie, chemical interactions among species, physical variations, temperature variations, humidity variations, spectrometer drift, changes in spectrometers, and insertion effects.

34. The method of Claim 25, wherein the vector comprises a set of random numbers.

35. A method of multivariate spectral analysis, comprising the steps of:

a) obtaining at least two reference variables for a first sample set comprised of at least one spectrally active component,

b) estimating at least one of the reference variables for the first sample set,

c) obtaining a residual between the at least one of the reference variables and its corresponding at least one estimated reference variable,

d) augmenting the at least one of the reference variables with a measure of the corresponding residual, and

e) predicting a value of at least one variable in a second sample set using the augmented reference variable obtained from the first sample.

36. The method of Claim 35, wherein the reference variables comprise spectra for the first sample set and component values of at least one component in the first sample set.

37. The method of Claim 35, wherein the augmentation step d) is repeated until all sources of spectral variation are accounted for in the first sample set.

38. The method of Claim 35, further comprising augmenting the at least one of the reference variables with at least one random number.

39. A method of multivariate spectral analysis, comprising the steps of:

a) obtaining an estimate of spectral error covariance $\mathbf{E_A}$ for measured set of multivariate spectral data $\mathbf{A}$;

b) decomposing the spectral error covariance $\mathbf{E_A}$ according to $\mathbf{E_A} = \mathbf{TP} + \mathbf{E}$, where $\mathbf{T}$ is a set of n x r scores and $\mathbf{P}$ is a set of r x p loading vectors obtained from factor analysis of the spectral error covariance $\mathbf{E_A}$, and $\mathbf{E}$ is a set of n x p random errors and spectral variations not useful for prediction;

c) guessing pure-component spectra $\mathbf{K}$ for the set of multivariate spectral data $\mathbf{A}$;

d) predicting a set of component values $\hat{\mathbf{C}}$ according to

$$\hat{\mathbf{C}} = \mathbf{AK}^{\mathrm{T}}(\mathbf{KK}^{\mathrm{T}})^{-1} = \mathbf{A}(\mathbf{K}^{\mathrm{T}})^{+};$$

e) augmenting the set of predicted component values $\hat{\mathbf{C}}$ with at least one vector of the $\mathbf{T}$ scores to obtain a first set of augmented component values $\hat{\tilde{\mathbf{C}}}$;

f) estimating augmented pure-component spectra $\hat{\tilde{\mathbf{K}}}$ according to

$$\hat{\tilde{\mathbf{K}}} = (\hat{\tilde{\mathbf{C}}}^{\mathrm{T}}\hat{\tilde{\mathbf{C}}})^{-1}\hat{\tilde{\mathbf{C}}}^{\mathrm{T}}\mathbf{A} = \hat{\tilde{\mathbf{C}}}^{+}\mathbf{A};$$

g) testing for convergence according to $\left\|\mathbf{A} - \hat{\tilde{\mathbf{C}}}\hat{\tilde{\mathbf{K}}}\right\|^2$;

h) predicting a second set of augmented component values $\hat{\tilde{\mathbf{C}}}$ according to

$$\hat{\tilde{\mathbf{C}}} = \mathbf{A}\hat{\tilde{\mathbf{K}}}^{\mathrm{T}}(\hat{\tilde{\mathbf{K}}}\hat{\tilde{\mathbf{K}}}^{\mathrm{T}})^{-1} = \mathbf{A}(\hat{\tilde{\mathbf{K}}}^{\mathrm{T}})^{+};$$

i) replacing the augmented portion of the second set of augmented component values $\hat{\tilde{\mathbf{C}}}$ with the at least one vector of the $\mathbf{T}$ scores to obtain a third set of augmented component values $\hat{\tilde{\mathbf{C}}}$; and

j) repeating steps f) through i) at least once.

40. The method of Claim 39, wherein the steps f) through i) are repeated until the test of step g) converges to obtain an alternating classical least squares solution for $\hat{\tilde{\mathbf{K}}}$ and $\hat{\tilde{\mathbf{C}}}$.

41. The method of Claim 39, further comprising replacing the augmented portion of the augmented pure-component spectra $\hat{\tilde{\mathbf{K}}}$ with at least one vector of the **P** loading vectors prior to step h).

42. The method of Claim 39, further comprising augmenting $\hat{\tilde{\mathbf{K}}}$ with at least one vector representing a spectral shape that is representative of at least one additional source of spectral variation prior to step h).

43. The method of Claim 39, further comprising applying at least one constraint to the non-augmented portion of $\hat{\tilde{\mathbf{K}}}$ at step f).

44. The method of Claim 43, wherein the at least one constraint is selected from the group consisting of non-negativity, equality, closure, monotonic constraint, unimodality, and selectivity.

45. The method of Claim 39, further comprising applying at least one constraint to the non-augmented portion of $\hat{\tilde{\mathbf{C}}}$ at step h).

46. The method of Claim 45, wherein the at least one constraint is selected from the group consisting of non-negativity, equality, closure, monotonic constraint, unimodality, and selectivity.

47. The method of Claim 39, wherein the guessed pure-component spectra **K** comprises random numbers.

48. The method of Claim 39, wherein the measured set of multivariate spectral data **A** comprises image data.

49. The method of Claim 48, wherein the spectral error covariance $\mathbf{E_A}$ is obtained from a shift difference generated from a single image.

50. The method of Claim 48, wherein the spectral error covariance $\mathbf{E_A}$ is obtained from repeat image spectra.

51. A method of multivariate spectral analysis, comprising the steps of:

a) obtaining an estimate of spectral error covariance $\mathbf{E_A}$ for measured set of multivariate spectral data $\mathbf{A}$;

b) decomposing the spectral error covariance $\mathbf{E_A}$ according to $\mathbf{E_A} = \mathbf{TP} + \mathbf{E}$, where $\mathbf{T}$ is a set of n x r scores and $\mathbf{P}$ is a set of r x p loading vectors obtained from factor analysis of the spectral error covariance $\mathbf{E_A}$, and $\mathbf{E}$ is a set of n x p random errors and spectral variations not useful for prediction;

c) guessing pure-component spectra $\mathbf{K}$ for the set of multivariate spectral data $\mathbf{A}$;

d) augmenting the pure-component spectra $\mathbf{K}$ with at least one vector of the $\mathbf{P}$ loading vectors to obtain first augmented pure-component spectra $\tilde{\mathbf{K}}$;

e) predicting a first set of augmented component values $\hat{\tilde{\mathbf{C}}}$ according to $\hat{\tilde{\mathbf{C}}} = \mathbf{A}\tilde{\mathbf{K}}^{\mathrm{T}}(\tilde{\mathbf{K}}\tilde{\mathbf{K}}^{\mathrm{T}})^{-1} = \mathbf{A}(\tilde{\mathbf{K}}^{\mathrm{T}})^{+}$;

f) estimating second augmented pure-component spectra $\hat{\tilde{\mathbf{K}}}$ according to $\hat{\tilde{\mathbf{K}}} = (\hat{\tilde{\mathbf{C}}}^{\mathrm{T}}\hat{\tilde{\mathbf{C}}})^{-1}\hat{\tilde{\mathbf{C}}}^{\mathrm{T}}\mathbf{A} = \hat{\tilde{\mathbf{C}}}^{+}\mathbf{A}$;

g) testing for convergence according to $\left\|\mathbf{A} - \hat{\tilde{\mathbf{C}}}\hat{\tilde{\mathbf{K}}}\right\|^{2}$;

h) replacing the augmented portion of the second augmented pure-component spectra $\hat{\tilde{\mathbf{K}}}$ with the at least one vector of the $\mathbf{P}$ loading vectors to obtain third augmented pure-component spectra $\hat{\tilde{\mathbf{K}}}$; and

i) predicting a second set of augmented component values $\hat{\tilde{\mathbf{C}}}$ according to $\hat{\tilde{\mathbf{C}}} = \mathbf{A}\hat{\tilde{\mathbf{K}}}^{\mathrm{T}}(\hat{\tilde{\mathbf{K}}}\hat{\tilde{\mathbf{K}}}^{\mathrm{T}})^{-1} = \mathbf{A}(\hat{\tilde{\mathbf{K}}}^{\mathrm{T}})^{+}$;

j) repeating steps f) through i) at least once.

52. The method of Claim 51, wherein the steps f) through i) are repeated until the test of step g) converges to obtain an alternating classical least squares solution for $\hat{\tilde{\mathbf{K}}}$ and $\hat{\tilde{\mathbf{C}}}$.

53. The method of Claim 51, further comprising replacing the augmented portion of the set of augmented component values $\hat{\tilde{\mathbf{C}}}$ with at least one vector of the $\mathbf{T}$ scores prior to step h).

54. The method of Claim 51, further comprising augmenting $\hat{\tilde{\mathbf{K}}}$ with at least one vector representing a spectral shape that is representative of at least one additional source of spectral variation prior to step h).

55. The method of Claim 51, further comprising applying at least one constraint to the non-augmented portion of $\hat{\tilde{\mathbf{K}}}$ at step f).

56. The method of Claim 55, wherein the at least one constraint is selected from the group consisting of non-negativity, equality, closure, monotonic constraint, unimodality, and selectivity.

57. The method of Claim 51, further comprising applying at least one constraint to the non-augmented portion of $\hat{\tilde{\mathbf{C}}}$ at step h).

58. The method of Claim 57, wherein the at least one constraint is selected from the group consisting of non-negativity, equality, closure, monotonic constraint, unimodality, and selectivity.

59. The method of Claim 51, wherein the guessed pure-component spectra $\mathbf{K}$ comprises random numbers.

60. The method of Claim 51, wherein the measured set of multivariate spectral data $\mathbf{A}$ comprises image data.

61. The method of Claim 60, wherein the estimate of the error covariance $\mathbf{E_A}$ is obtained from a shift difference generated from a single image.

62. The method of Claim 60, wherein the estimate of the error covariance $\mathbf{E_A}$ is obtained from repeat image spectra.

63. A method of multivariate spectral analysis, comprising the steps of:

a) obtaining an estimate of the spectral error covariance $\mathbf{E_A}$ for measured set of multivariate spectral data $\mathbf{A}$;

b) decomposing the spectral error covariance $\mathbf{E_A}$ according to $\mathbf{E_A} = \mathbf{TP} + \mathbf{E}$, where $\mathbf{T}$ is a set of n x r scores and $\mathbf{P}$ is a set of r x p loading vectors obtained from factor analysis of the spectral error covariance $\mathbf{E_A}$, and $\mathbf{E}$ is a set of n x p random errors and spectral variations not useful for prediction;

c) guessing a set of component values $\mathbf{C}$ for the set of multivariate spectral data $\mathbf{A}$;

d) estimating pure-component spectra $\hat{\mathbf{K}}$ according to $\hat{\mathbf{K}} = (\mathbf{C}^T\mathbf{C})^{-1}\mathbf{C}^T\mathbf{A} = \mathbf{C}^+\mathbf{A}$;

e) augmenting the pure-component spectra $\hat{\mathbf{K}}$ with at least one vector of the $\mathbf{P}$ loading vectors to obtain first augmented pure-component spectra $\hat{\tilde{\mathbf{K}}}$;

f) predicting a first set of augmented component values $\hat{\tilde{\mathbf{C}}}$ according to $\hat{\tilde{\mathbf{C}}} = \mathbf{A}\hat{\tilde{\mathbf{K}}}^T(\hat{\tilde{\mathbf{K}}}\hat{\tilde{\mathbf{K}}}^T)^{-1} = \mathbf{A}(\hat{\tilde{\mathbf{K}}}^T)^+$;

g) testing for convergence according to $\left\|\mathbf{A} - \hat{\tilde{\mathbf{C}}}\hat{\tilde{\mathbf{K}}}\right\|^2$;

h) estimating second augmented pure-component spectra $\hat{\tilde{\mathbf{K}}}$ according to $\hat{\tilde{\mathbf{K}}} = (\hat{\tilde{\mathbf{C}}}^T\hat{\tilde{\mathbf{C}}})^{-1}\hat{\tilde{\mathbf{C}}}^T\mathbf{A} = \hat{\tilde{\mathbf{C}}}^+\mathbf{A}$;

i) replacing the augmented portion of the second augmented pure-component spectra $\hat{\tilde{\mathbf{K}}}$ with the at least one vector of the $\mathbf{P}$ loading vectors to obtain a third augmented pure-component spectra $\hat{\tilde{\mathbf{K}}}$ and

j) repeating steps f) through i) at least once.

64. The method of Claim 63, wherein the steps f) through i) are repeated until the test of step g) converges to obtain an alternating classical least squares solution for $\hat{\tilde{\mathbf{K}}}$ and $\hat{\tilde{\mathbf{C}}}$.

65. The method of Claim 63, further comprising replacing the augmented portion of the set of augmented component values $\hat{\tilde{\mathbf{C}}}$ with at least one vector of the **T** scores prior to step h).

66. The method of Claim 63, further comprising augmenting $\hat{\tilde{\mathbf{K}}}$ with at least one vector representing a spectral shape that is representative of at least one additional source of spectral variation prior to step h).

67. The method of Claim 63, further comprising applying at least one constraint to the non-augmented portion of $\hat{\tilde{\mathbf{K}}}$ at step h).

68. The method of Claim 67, wherein the at least one constraint is selected from the group consisting of non-negativity, equality, closure, monotonic constraint, unimodality, and selectivity.

69. The method of Claim 63, further comprising applying at least one constraint to the non-augmented portion of $\hat{\tilde{\mathbf{C}}}$ at step f).

70. The method of Claim 69, wherein the at least one constraint is selected from the group consisting of non-negativity, equality, closure, monotonic constraint, unimodality, and selectivity.

71. The method of Claim 63, wherein the guessed set of component values **C** comprises random numbers.

72. The method of Claim 52, wherein the measured set of multivariate spectral data **A** comprises image data.

73. The method of Claim 62, wherein the spectral error covariance $\mathbf{E_A}$ is obtained from a shift difference generated from a single image.

74. The method of Claim 62, wherein the spectral error covariance $\mathbf{E_A}$ is obtained from repeat image spectra.

75. A method of multivariate spectral analysis, comprising the steps of:

a) obtaining an estimate of the spectral error covariance $\mathbf{E_A}$ for measured set of multivariate spectral data $\mathbf{A}$;

b) decomposing the spectral error covariance $\mathbf{E_A}$ according to $\mathbf{E_A} = \mathbf{TP} + \mathbf{E}$, where $\mathbf{T}$ is a set of n x r scores and $\mathbf{P}$ is a set of r x p loading vectors obtained from factor analysis of the spectral error covariance $\mathbf{E_A}$, and $\mathbf{E}$ is a set of n x p random errors and spectral variations not useful for prediction;

c) guessing a set of component values $\mathbf{C}$ for the set of multivariate spectral data $\mathbf{A}$;

d) augmenting the set of component values $\mathbf{C}$ with at least one vector of the $\mathbf{T}$ scores to obtain a first set of augmented component values $\tilde{\mathbf{C}}$;

e) estimating augmented pure-component spectra $\hat{\tilde{\mathbf{K}}}$ according to $\hat{\tilde{\mathbf{K}}} = (\tilde{\mathbf{C}}^T\tilde{\mathbf{C}})^{-1}\tilde{\mathbf{C}}^T\mathbf{A} = \tilde{\mathbf{C}}^+\mathbf{A}$;

f) testing for convergence according to $\left\|\mathbf{A} - \tilde{\mathbf{C}}\hat{\tilde{\mathbf{K}}}\right\|^2$;

g) predicting a second set of augmented component values $\hat{\tilde{\mathbf{C}}}$ according to $\hat{\tilde{\mathbf{C}}} = \mathbf{A}\hat{\tilde{\mathbf{K}}}^T(\hat{\tilde{\mathbf{K}}}\hat{\tilde{\mathbf{K}}}^T)^{-1} = \mathbf{A}(\hat{\tilde{\mathbf{K}}}^T)^+$;

h) replacing the augmented portion of the second set of augmented component values $\hat{\tilde{\mathbf{C}}}$ with the at least one vector of the $\mathbf{T}$ scores to obtain a third set of augmented component values $\hat{\tilde{\mathbf{C}}}$ and

i) repeating steps e) through h) at least once, using the augmented component values $\hat{\tilde{\mathbf{C}}}$ in step f).

76. The method of Claim 75, wherein the steps e) through h) are repeated until the test of step g) converges to obtain an alternating classical least squares solution for $\hat{\tilde{\mathbf{K}}}$ and $\hat{\tilde{\mathbf{C}}}$.

77. The method of Claim 75, further comprising replacing the augmented portion of the augmented pure-component spectra $\hat{\tilde{\mathbf{K}}}$ with at least one vector of the $\mathbf{P}$ loading vectors prior to step e).

78. The method of Claim 75, further comprising augmenting $\hat{\tilde{\mathbf{K}}}$ with at least one vector representing a spectral shape that is representative of at least one additional source of spectral variation prior to step e).

79. The method of Claim 75, further comprising applying at least one constraint to the non-augmented portion of $\hat{\tilde{\mathbf{K}}}$ at step e).

80. The method of Claim 79, wherein the at least one constraint is selected from the group consisting of non-negativity, equality, closure, monotonic constraint, unimodality, and selectivity.

81. The method of Claim 75, further comprising applying at least one constraint to the non-augmented portion of $\hat{\tilde{\mathbf{C}}}$ at step g).

82. The method of Claim 81, wherein the at least one constraint is selected from the group consisting of non-negativity, equality, closure, monotonic constraint, unimodality, and selectivity.

83. The method of Claim 75, wherein the guessed set of component values $\mathbf{C}$ comprises random numbers.

84. The method of Claim 75, wherein the measured set of multivariate spectral data $\mathbf{A}$ comprises image data.

85. The method of Claim 84, wherein the spectral error covariance $\mathbf{E_A}$ is obtained from a shift difference generated from a single image.

86. The method of Claim 84, wherein the spectral error covariance $\mathbf{E_A}$ is obtained from repeat image spectra.